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Signed Andrew Gersey

Dated 14 December 1999

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Request for grant of a patent

(See the notes on the back of this form. You can also get an explanatory leaflet from the Patent Office to help you fill in this form)

The Patent Office

Cardiff Road
Newport
Gwent NP9 1RH

1. Your reference

JRL 1437 GB

2. Patent application number
(The Patent Office will fill in this part)

03 DEC 1998 9826486.4

3. Full name, address and postcode of the or of each applicant *(underline all surnames)*

JOHNSON MATTHEY PUBLIC LIMITED COMPANY
2-4 COCKSPUR STREET
TRAFALGAR SQUARE
LONDON SW1Y 5BQ

Patents ADP number *(if you know it)*

536268007

If the applicant is a corporate body, give the country/state of its incorporation

GB

4. Title of the invention

IMPROVEMENTS IN COATINGS

5. Name of your agent *(if you have one)*

IAN CARMICHAEL WISHART

"Address for service" in the United Kingdom to which all correspondence should be sent *(including the postcode)*

JOHNSON MATTHEY TECHNOLOGY CENTRE
BLOUNTS COURT
SONNING COMMON
READING RG4 9NH

7258312001

Patents ADP number *(if you know it)*

6. If you are declaring priority from one or more earlier patent applications, give the country and the date of filing of the or of each of these earlier applications and *(if you know it)* the or each application number

Country	Priority application number *(if you know it)*	Date of filing *(day / month / year)*

7. If this application is divided or otherwise derived from an earlier UK application, give the number and the filing date of the earlier application

Number of earlier application	Date of filing *(day / month / year)*

8. Is a statement of inventorship and of right to grant of a patent required in support of this request? *(Answer 'Yes' if:*
a) any applicant named in part 3 is not an inventor, or
b) there is an inventor who is not named as an applicant, or
c) any named applicant is a corporate body.
See note (d))

Yes

Patents Form 1/77

9. Enter the number of sheets for any of the following items you are filing with this form. Do not count copies of the same document

Continuation sheets of this form	0
Description	4
Claim(s)	0
Abstract	0
Drawing(s)	1 +1

10. If you are also filing any of the following, state how many against each item.

Priority documents

Translations of priority documents

Statement of inventorship and right to grant of a patent *(Patents Form 7/77)*

Request for preliminary examination and search *(Patents Form 9/77)*

Request for substantive examination *(Patents Form 10/77)*

Any other documents *(please specify)*

11. I/We request the grant of a patent on the basis of this application.

Signature [signature] Date 2 Dec 98

I C WISHART

12. Name and daytime telephone number of person to contact in the United Kingdom

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Case JRL 1437

IMPROVEMENTS IN COATINGS

This invention concerns improvements in coatings, and more especially concerns improvements in methods of coating metal oxides and the like onto metallic or non-metallic substrates.

It is very well established to coat metal oxides, such as alumina, ceria, zirconia, etc, as single or mixed oxides, onto metal substrates and ceramic such as honeycomb through-flow catalyst supports, used in huge numbers in exhaust gas catalysts. This is generally achieved by admixing the metal oxide into an aqueous slurry together with suitable components including adhesion modifiers, of which the "Ludox" (Trade Mark) silica is an example, to form a washcoat slurry. The substrate is then dipped or drenched in the slurry, and fired to yield a high surface area base for the catalyst. The silica may form 50 wt% or more of the solids content of the slurry, and this clearly dilutes the interaction between the catalyst metals and the oxide, which is very important. There has not been significant improvement in these coating techniques for some 20 or 30 years, yet the design of exhaust gas catalysts is becoming more sophisticated and demanding, requiring new components such as zeolites and other multi-layers to be incorporated. We believe that traditional coating methods are failing to deal with new requirements, and in particular, traditional coating methods give problems in multi-layer coating.

We have previously disclosed the use of polyelectrolytes to coat catalyst particles etc with zeolites formed simultaneously *in situ* (see EP 878 233). We have now discovered that polyelectrolytes can be used to pre-treat metal and ceramic substrates without such *in situ* formation of the zeolites, in a process in which one or more pre-formed metal oxides, including zeolites and the like, is deposited onto the substrate, and not only is a sound, even coating obtained without requiring dilution with adhesion modifiers, (but may include such adhesion modifiers) but the coating is especially suitable for the deposition of further coatings of similar or differing type.

Accordingly, the present invention provides a method of depositing metal oxide coatings onto a substrate, which comprises treating the surface of said substrate with a polyelectrolyte to yield a surface coating of the polyelectrolyte thereon, and subsequently or simulaneously treating the coated surface with a an aqueous slurry comprising metal oxide particles. If required, additional layers of the same or different metal oxide may be applied, either to increase the loading of the metal oxide or to form a multilayer coating.

Desirably, the polyelectrolyte is applied in the form of an aqueous solution, eg a 0.01 to 20 wt%, in the case of the materials specifically described hereinafter a 4 wt%, solution has been found satisfactory. The polyelectrolyte may be any suitable polyelectrolyte, eg anionic or cationic, but it is presently preferred to use a polyacrylamide, such as PE-1679 available from Allied Colloids Ltd, England. For such materials, desirably, the solution is alkaline, for example of pH of approximately 9. Routine testing to optimise the solution pH for each substrate and polyelectrolyte should be undertaken, and may be acid or alkaline, primarily depending upon the surface chemistry of the substrate. Treatment of the substrate may be by any suitable method, including spraying, dipping, vacuum application, drenching using a "waterfall" and the like, and may be carried out at room temperature. Desirably, the polyelectrolyte solution is dried to form a continuous surface polymer layer.

The substrate may be any metal or ceramic material, in any form, including particularly honeycomb through-flow catalyst supports, but also devices of the type known as static mixers, which provide good gas or other fluid mixing. The metal may be, for example, a stainless steel, including "Fecralloy" or aluminium, and the ceramic may be cordierite or the like. Other substrates which require, or may be protected by, an oxide coating, should also be considered.

The metal oxide includes zeolites of all types and of all Si to Al ratios, and includes modified, eg ion-exchanged zeolites, and single or mixed oxides, for example selected from one or more of ceria, zirconia, magnesia, alumina and silica. The metal oxide is desirably in the form of an aqueous slurry, eg of about 40 wt% solids content. The slurry may, but need not, include an adhesion

modifier, and may include other components including catalytically active particles and solutions of catalytically active metals and/or promoters therefor.

The slurry is then suitably applied by any method to the polyelectrolyte-coated substrate, and is then suitably dried. A final stage is desirably firing, or calcining, to firmly deposit the oxide coating on the substrate. Before the final firing, however, additional slurries of oxide and/or other components may be deposited, to result in a continuous or thicker coating, or a multi-component series of coatings. If desired or necessary, a further polyelectrolyte layer may be applied, and one or more additional metal oxide or other coatings may be applied.

In an alternative embodiment, the polyelectrolyte is admixed with the slurry without a separate pre-coating step. Otherwise the process and materials considerations are fairly similar. This embodiment is expected to be particularly suitable for depositing a thin, even coating.

It is believed, although we do not wish to be bound by any theory, that the polelectrolyte acts to reverse the charge on the substrate, thus permitting the slurry particles to adhere firmly, and we believe that upon deposition of a second coating, the polyelectrolyte is "re-activated" and serves to cause excellent adhesion of the second coating.

It is believed that the present invention has significant and unexpected benefits in achieving good coatings on substrates that have previously been difficult to coat, and in particular permits sound and adherent two or multi-layer coatings.

The present invention is illustrated by the following example.

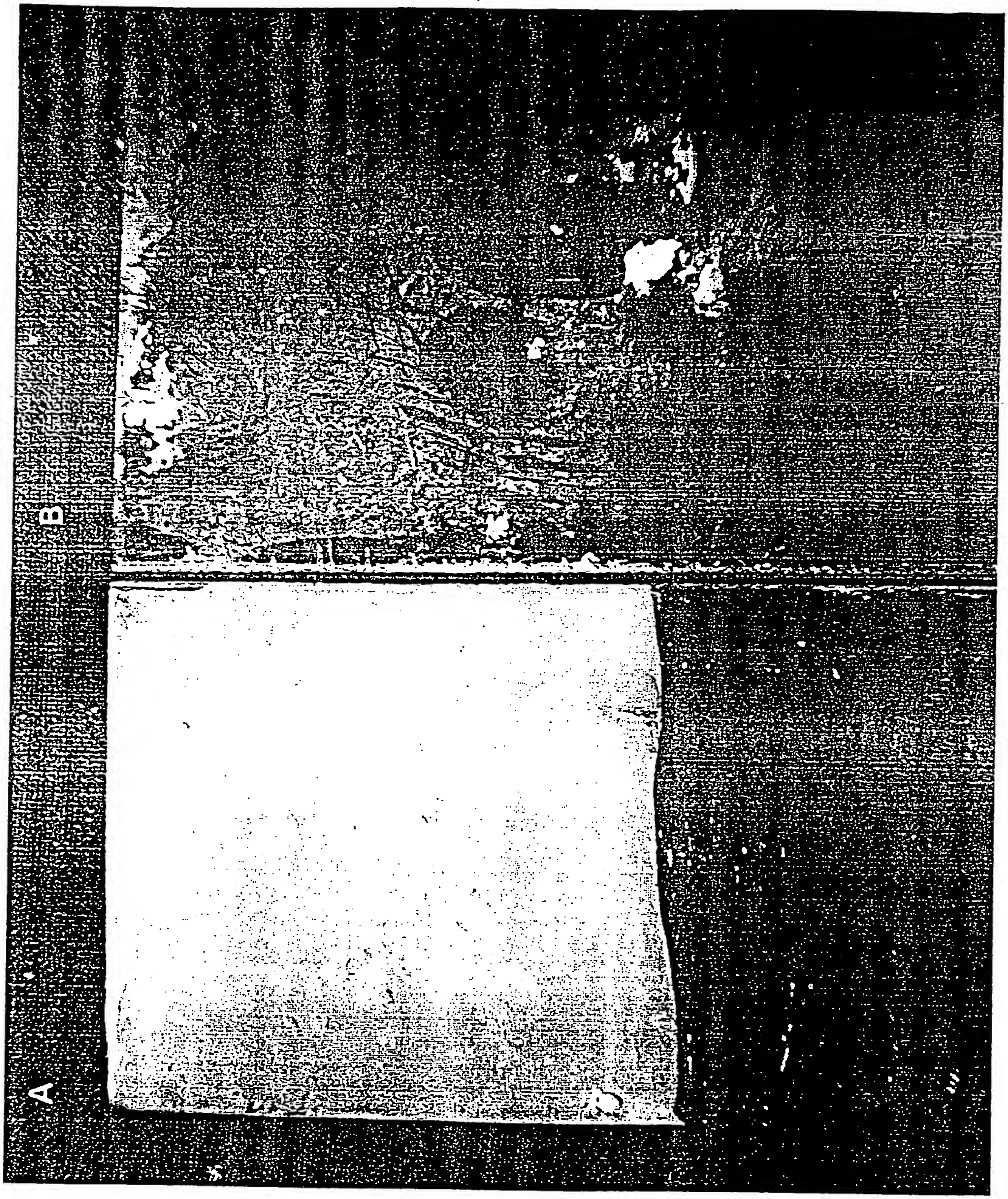
EXAMPLE 1

An aluminium plate is soaked with aqueous 4 wt% polyacrylamide solution at pH 9. The wet plate is then dried to leave a thin surface coating of polyelectrolyte. The coated plate is dipped into a

standard zeolite washcoat slurry as used in the exhaust gas catalyst industry, containing 40 wt% zeolite, with the pH adjusted to between 8 and 9. The washcoat adheres to the plate, and the coated plate is removed and dried at 100 °C for 30 minutes. A further identical washcoat layer was then applied to deposit a desired thickness. The coated metal plate is then calcined at 400°C for 2 hours, which removes the polyelectrolyte residue. A photograph of the thus-coated plate ("A") is shown in the accompanying drawing page. For comparison, the identical procedure was followed without the polyelectrolyte treatment. Although a first coating appeared successful and even, a second coating caused the combined coating to dramatically peel; a photograph of the coated plate is shown as "B" in the drawing. It can readily be seen that the present invention permits an even and continuous coating, whereas double coating without the polyelectrolyte results in flaking of the coating, and uneven, discontinuous coating.

Although the above Example utilised a 100% zeolite coating, other successful coatings can be applied with 50 wt% "Ludox" silica adhesion modifier, or any other proportion.

The invention may be modified by the skilled person without departing from the scope thereof.


A
B

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